ARTICLE IN PRESS

Journal of Crystal Growth ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at ScienceDirect

Journal of Crystal Growth

journal homepage: www.elsevier.com/locate/jcrysgro

Impact of *in-situ* annealing on dilute-bismide materials and its application to photovoltaics

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ARTICLE INFO

Keywords:
A1. High resolution X-ray diffraction
A3. Organometallic vapor phase epitaxy
B2. Semiconducting III–V materials
B2. Semiconducting ternary compounds
B3. Solar cells

ABSTRACT

$GaAs_{1-x}Bi_x$/GaAs multiple quantum well heterostructures were grown by organo-metallic vapor phase epitaxy (OMVPE) at low temperatures and were subsequently *in-situ* annealed under an arsine (AsH_3) overpressure in the OMVPE reactor. Photoluminescence (PL) measurements were performed to establish the optimized annealing condition for the highest luminescence intensity, as well as high resolution X-ray diffraction (XRD) measurements to detect any structural changes after annealing. In addition, the complex compositional profile and the interfacial abruptness were deduced from the combined XRD analysis with the transmission electron microscopy. A 5-fold increase in the PL intensity at room temperature was observed after annealing under the optimized conditions. Using the optimized annealing conditions, single junction solar cells (SJSC) incorporating 5-period $GaAs_{0.964}Bi_{0.036}$/GaAs (32 nm/20 nm) heterostructures for the device base region were fabricated and characterized. The spectral dependence of the external quantum efficiency of the SJSC exhibited improved spectral response as a result of the optimized *in-situ* annealing, with extended absorption edge up to 1.07 eV.

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1. Introduction

Recently, $GaAs_{1-x}Bi_x$ alloys have been drawing much attention since it was shown both experimentally and theoretically that a small amount of Bi incorporation into GaAs can induce a rapid reduction in the band gap energy (E_g) by 60–80 meV per %Bi in $GaAs_{1-x}Bi_x$ [1–5]. The origin of the rapid band gap reduction has been attributed to the valence band anti-crossing from the localized interaction between the Bi resonant energy level and the valence band of GaAs, which results in a relatively small conduction band offset between GaAs and $GaAs_{1-x}Bi_x$ for small Bi fractions [6].

However, there are several challenges to utilize $GaAs_{1-x}Bi_x$ material system for optoelectronic devices. First, $GaAs_{1-x}Bi_x$ is a metastable alloy system requiring its growth to be carried out far away from thermodynamic equilibrium at a very low growth temperature (~380–420 °C) in order to incorporate a sufficient amount of Bi into the GaAs host matrix. Such low growth temperatures may lead to degradation in radiative efficiency of the as-grown material, similar to that observed in the dilute-nitride material system [7]. Our previous study indicated *in-situ* annealing under an arsine overpressure can dramatically increase the luminescence intensity, evidenced by low temperature (15 K) PL, of $GaAs_{0.97}Bi_{0.03}$ single quantum wells employing tensile-strained $GaAs_{0.7}P_{0.3}$ barriers [8]. Here, we report a more comprehensive study on the effect of *in-situ* annealing on the luminescence and structural properties of $GaAs_{1-x}Bi_x$/GaAs grown by OMVPE, which is a potential candidate as a MQW base region for the 1 eV middle cell in a multi-junction solar cell.

Also, a complex bismuth distribution in a Bi-containing layer has been reported by Forghani et al. as a result of the relatively long surface residence time of Bi precursors or the reaction products blocking surface sites for the adsorption and dissociation of the group III source [9]. We observe a complex Bi distribution within the relatively thick (~32 nm) $GaAs_{1-x}Bi_x$ layers of the $GaAs_{1-x}Bi_x$/GaAs MQW by means of the transmission electron microscopy.

To date, there have been a few studies on the utilization of the $GaAs_{1-x}Bi_x$ material system as the solar cell base region material [10], although several demonstrations for light emitting devices have been shown [11–14]. For the photovoltaic application, when grown on a GaAs substrate, the $GaAs_{1-x}Bi_x$ material system has

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http://dx.doi.org/10.1016/j.jcrysgro.2016.04.039
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Please cite this article as: H. Kim, et al., Journal of Crystal Growth (2016), http://dx.doi.org/10.1016/j.jcrysgro.2016.04.039

the advantage over the more conventional $In_zGa_{1-z}As$ material system in that a smaller compressive strain is induced at the same band gap energy, allowing a much thicker $GaAs_{1-x}Bi_x$ to be employed without encountering strain relaxation; thus leading to increased optical absorption from a thicker solar cell base region. Finally, the demonstration of single junction solar cells (SJSC) employing $GaAs_{1-x}Bi_x$ as a base region material is presented here, showing the *in-situ* annealing can lead to a significant improvement in the device performance.

2. Experimental procedure

A sample for annealing study, a 5-period $GaAs_{0.964}Bi_{0.036}$/GaAs (32/20 nm), was grown by OMVPE on exact (100) semi-insulating GaAs substrates as shown in Fig. 1, using Pd-purified H_2 as a carrier gas at 76 T. The $GaAs_{1-x}Bi_x$ was grown by the simultaneous feed of triethyl gallium (TEGa), triethyl bismuth (TEBi), and tertiarybutyl arsine (TBAs) at the growth temperature of 380 °C while the GaAs barriers were grown using TEGa and arsine (AsH_3) at the elevated temperature of 650 °C after the $GaAs_{0.964}Bi_{0.036}$ well growth was paused as summarized in Table 1. The effects of these specific precursors on the growth of $GaAs_{1-x}Bi_x$ were discussed elsewhere [9]. The growth interruption and temperature ramp-up are thought to evaporate the segregated Bi, which remains on the surface of bismide-containing well layer, as reported elsewhere [11,15]. The temperature ramp-up from 380 °C and stabilization at 650 °C took approximately 3 min. After the growth, samples were cleaved into multiple pieces and annealed in the OMVPE reactor at various temperatures under the stabilized arsine ambient (4900 μmol/min) for 40 min, which also represents a typical time taken for the growth of the upper device structure layers such as the emitter, window and contact layers for the single junction solar cell structure. Room Temperature PL (RT-PL) measurements were performed to investigate the impact of the post-growth thermal annealing on the luminescence properties, using continuous wave Nd:YAG laser at the excitation wavelength of 532 nm, 300 g/mm grating monochromator, and cooled InGaAs detector. The HR-XRD $\omega-2\theta$ scan around the (004) reflection was carried out to detect any significant structural change in the annealed samples. The average Bi fraction and thickness were determined by the HR-XRD pattern, with the assumption of 0.633 nm as the GaBi lattice constant and with curve fitting of the zero-order superlattice peak position [4]. Along with the HR-XRD measurement, the high-angle annular dark-field (HAADF), or "z-contrast", imaging method in an aberration corrected scanning transmission electron microscopy (STEM) measurement was used to investigate the spatial distribution of Bi incorporated into GaAs during the growth as well as the interfacial abruptness at the $GaAs_{1-x}Bi_x$/GaAs hetero-interfaces. Profiles of the STEM image intensity along lines extending from the GaAs buffer layer were generated in the same way as described in Ref. [16]. The cross-sectional STEM sample was prepared by the wedge-polishing method, followed by Ar ion milling.

Table 1
Key growth parameters employed in the OMVPE material growth.

Material	Molar flow (μmol/min)					Growth Temperature (°C)	Growth Rate (nm/s)
	TEGa	TBAs	TEBi	AsH_3	PH_3		
$GaAs_{0.964}Bi_{0.036}$	57	145	0.6	–	–	380	0.12
GaAs	13	–	–	3800	–	650	0.25

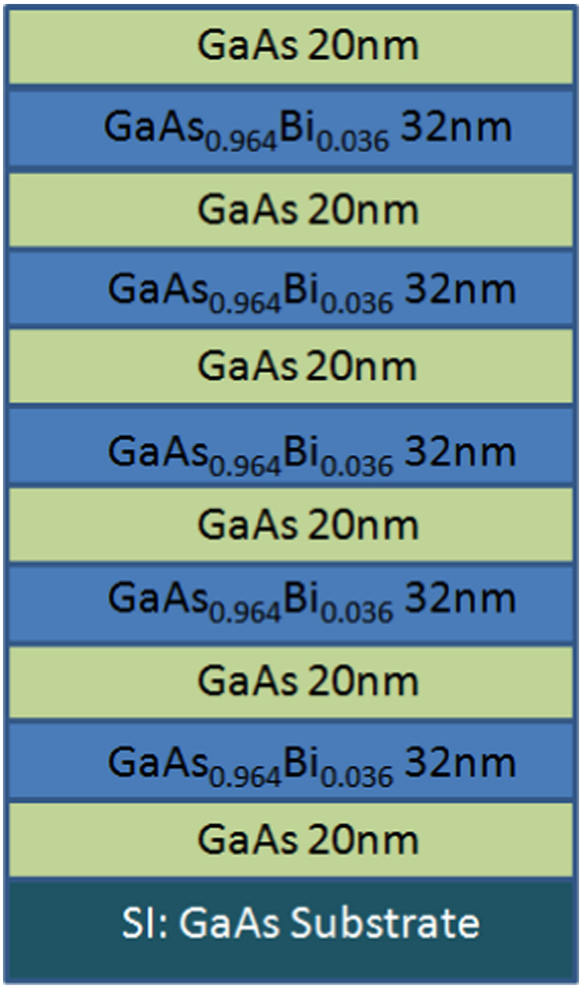

Fig. 1. Structure details for the samples employed in the annealing study. GaAs was grown at 650 °C while $GaAs_{0.964}Bi_{0.036}$ was grown at 380 °C. Bi mole fraction is an average value in a $GaAs_{1-x}Bi_x$ layer deduced from the HR-XRD measurement.

Two SJSC structures employing 5 periods of $GaAs_{0.964}Bi_{0.036}$/GaAs (32/20 nm) as a base region were also grown and subjected to thermal annealing during the growth of the emitter, the window, and contact layers above the base region. After the material growth, solar cells were fabricated with an active area of 0.2 cm^2 defined by the illuminated region between metal bonding pads. Ohmic contacts to n-and p-GaAs were made using AuGe/Ni/Au and Ti/Pt/Au metallurgy, followed by the rapid thermal annealing at 385 °C.

3. Results and discussion

First, for the annealing study, a 5 period $GaAs_{0.964}Bi_{0.036}$/GaAs (32/20 nm) sample grown on semi-insulating GaAs substrate was annealed at various temperatures, 650, 700, and 800 °C, for 40 min under an arsine/hydrogen ambient. The indicated Bi mole fraction is an average value over a Bi-containing layer deduced from the HR-XRD measurement assuming the uniform Bi distribution. The sample annealed at 650 °C exhibited a 5-fold increase in PL intensity at room temperature compared to that of the as-grown materials, with the peak position at 1.08 μm (Fig. 2a). No structural change was evident from the $\omega-2\theta$ HR-XRD pattern after annealing at 650 °C although the higher temperature annealing at 700 °C resulted in the degraded peak-to-valley contrast in the pendellÖsung fringes, indicating the roughened interface at $GaAs_{0.964}Bi_{0.036}$/GaAs heterojunction. In addition, annealing at 800 °C caused the out-diffusion of Bi as evident from HR-XRD $\omega-2\theta$ scan in Fig. 2b. It should be noted that no obvious spectral peak shift in the PL was observed after the post-growth thermal

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